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A Keynote Talk

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Publication date:
2019

Document Version
Publisher's PDF, also known as Version of record

Link to publication from Aalborg University

Citation for published version (APA):
Yue, Y., Zhang, Y. F., Tao, H. Z., Qiao, A., & Xiong, F. Y. (2019). *Enhancing the Li/Na-Ion Battery Performances by Disorder/Order Engineering: A Keynote Talk*. Abstract from Angell International Symposium, Peyia, Cyprus.

Enhancing the Li/Na-Ion Battery Performances by Disorder/Order Engineering

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We are facing big challenges in developing full-solid Li/Na ion batteries concerning the limited performances and problems of electrodes and solid electrolytes. However, there are also potential possibilities to overcome these challenges. In this presentation, we demonstrate a different route, that is, our disorder/ordering engineering concept [1] to develop high performance cathode/anode/electrolyte materials. The disorder/order engineering refers to two aspects. **First**, part of the disordered or glass structure in cathode/anode materials is transformed into the ordered domains. **Second**, the long-range ordered solids are transformed into disordered or amorphous ones. In this talk, we present three case studies concerning the effect of the disorder/order engineering on the electrochemical performances of cathodes, anodes, and solid electrolytes, respectively, for Li/Na-ion batteries.

Case 1: A series of vanadium-tellurite glasses with various V/Te ratios were synthesized via melt-quenching [1,2], and then the glass was pulverized and mixed with carbon for making Li-ion battery anodes. The anodes underwent discharging/charging cycles. During cycling, a fascinating phenomenon was observed, i.e., nanocrystals formed in glass matrix. As a consequence, the cycling stability and electronic/ionic conductivity of the anodes were enhanced. This kind of nanocrystal formation has a fundamentally different origin compared to the thermally induced crystallization [1,3].

Case 2: $NaFePO_4$ with maricite structure, which is thermodynamically stable phase, was considered to be electrochemically inactive for sodium-ion storage. Recently, we succeeded in creating disorder in $NaFePO_4$ cathode by a mechanochemical route to enhance electrochemical performances of Na-ion batteries [4]. The derived $NaFePO_4$ cathodes containing both amorphous and maricite phases exhibit much improved sodium storage performance with an initial capacity of 115 mA h g^{-1} at 1 C and an excellent cycling stability of capacity retention of 91.3% after 800 cycles.

Case 3: The crystalline Ag_3PS_4 was transformed into amorphous state via a chemo-mechanical milling process. The Ag^+ conductivity of the amorphous sample was found to be about three orders of magnitude higher than that of the crystalline counterpart. The amorphous sample exhibits lower activation energy (E_a) for the Ag^+ migration, and hence, lower Ag^+ conductivity compared to the crystalline one. By performing structural characterizations, we explored the origin of the enhanced Ag^+ conductivity of the amorphous sample. The present study provided valuable information for developing solid electrolytes.

Keywords: Li/Na Ion Batteries; Order/Disorder Engineering; Glass; Cathode; Anode; Fast Ionic Conductors

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